REMARKS:

Favorable reconsideration is respectfully requested in view of the foregoing amendments and following remarks.

Claims 1 and 2 have been cancelled without prejudice and new claims 3-20 have been added. The new claims have been drafted in better conformance with U.S. practice. Specifically, claims 3-5 are supported by original claim 1. Claims 6 and 7 are supported by original claim 2. Claim 8 is supported in the specification at page 4, line 1. Claims 9-14 are supported by page 4 of the specification. Claims 15-20 are supported by page 1, lines 1-2, page 4, lines 2-5 and page 5.

Turning to the Official Action, there is submitted herewith a copy of EP 0034403 in reply to item 2 on page 2 of the Action. The two other cited references, GB 2144336 and US Patent 3,839,229, were previously submitted to the Examiner and have been considered by the Examiner, as reflected in the PTO-1449 form dated November 27, 2000 attached to the Office Action. Consideration of EP 0034403 is respectfully solicited.

The specification has been revised in conformance with U.S. practice.

The objection to the disclosure set forth in item 3 is submitted to be overcome.

An abstract has been added corresponding to the abstract published in the published international application.

The objection to the abstract is submitted to be overcome.

Lastly, claims and 1 and 2 were rejected under 35 U.S.C. 103 as obvious over Senes et al. in view of Burnett. This current rejection is respectfully traversed as applied to the new claims presented.

Senes et al. relates to an ammonia synthesis catalyst consisting of iron oxide and cobalt oxide. It may further include a promotor selected from the group consisting of alumina, silica, zirconia, magnesia, lime, potassium oxide and rare earth metal oxides. It is stated that the introduction of cobalt permits the obtaining of excellent catalysts, even in the absence of promoters. Cobalt oxide is present in an amount of 5-10 weight percent expressed as cobalt. This catalyst is said to have high activity at low temperatures (200-400°C). However, as recognized by the Examiner, Senes et al. fail to disclose or suggest a catalyst including titanium oxide according to the instant claims.

Burnett relates to an improved ammonia synthesis catalyst containing platinum. Under the description of prior art in column 1, lines 45-50, in the patent, it is referred to an article by H. Hinrichs in British Chemical Engineering, where several known metal oxide promoters are listed, among these also TiO_2. Cobalt oxide is not on the list. On lines 50-55 of column 1, the reference further states that "According to Dr. Hinrichs, who has worked on ammonia synthesis for more than 15 years, apart from Al_2O_3 and K_2O the only promoters of practical importance are MgO and SiO_2, and even as to these promoters Hinrichs states that they increase stability at high temperatures and have resistance to poisoning, **but at the penalty of reduced catalyst activity.**"

The main objective of the present invention is to develop a catalyst for ammonia synthesis with improved activity. Even if Burnett lists TiO_2 as a promotor, Applicant sees no suggestion or motivation in the reference to try titanium oxide in the combination with cobalt oxide. Firstly, cobalt oxide is not mentioned as a possible promoter. Secondly, the effect of the combination with titanium oxide could not be predicted. On the contrary, if an improved catalyst activity is the object, this reference teaches that many of the promoters reduce the catalyst activity.

In Senes, it is stated that the introduction of cobalt permits the obtaining of excellent catalysts, even in the absence of promoters. Thus, there should be no reason to try other promoters. Furthermore, note that the concentration of Co used according to Senes is much higher than in the present application, between 5 and 10 weight % expressed as cobalt compared to between 0.1 and 3 percent in the present application. Applicants obtain excellent results with a much lower content of Co in combination with Ti.

The teachings of the cited references do not suggest or motivate one of ordinary skill in the art to prepare a catalyst containing both cobalt oxide and titanium oxide.

In view of the foregoing, favorable reconsideration and allowance is respectfully solicited.

Respectfully submitted,

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BACKGROUND OF THE INVENTION

1. Field of the Invention

CATALYST FOR THE SYNTHESIS OF AMMONIA FROM HYDROGEN AND NITROGEN

The present invention relates to a catalyst for the synthesis of ammonia from hydrogen and nitrogen.

2. Description of Related Art

The ammonia synthesis catalyst play an important role in the manufacture of ammonia. It effects both the economics and operating conditions of ammonia plants.

Industrial catalysts for ammonia synthesis must satisfy a number of requirements: high catalyst activity at the lowest possible reaction temperatures in order to take advantage of the favourable thermodynamic equilibrium at low temperatures, good resistance to poisoning by oxygen-, chlorine- and sulphur -containing compounds, long life and high mechanical strength.

In order to save energy the synthesis pressure has been steadily reduced from 250-350 bar to 80-150 bar during the last 30 years due to optimised operation of the synthesis loop made possible by new technology, especially for fabrication of the converter. Improvements in the front end has drastically reduced the content of catalyst poisons (oxygen- , sulphur- and chlorine-containing compounds) in the gas inlet to the converter. However, the drop in synthesis pressure necessitates a three- to four-fold increase in the catalyst volume. The demands upon the catalyst quality has also increased. Apparently small improvements in the catalyst activity may lead to major improvements of the operation of a modern ammonia synthesis loop.

The ammonia synthesis catalyst precursor, the unreduced catalyst, is manufactured by melting iron oxides, mainly magnetite, and oxides or carbonates of Al, K, Ca and Mg, the so-called promoters. The melting is performed in electrical furnaces at a temperature of above 1600 °C. The ratio Fe^{2+}/Fe^{3+} in the melt is normally in the range 0.5-0.75. The melt is then poured into metal trays where it solidifies and cools. The

chemisorbed CO normally increase with promoter content, but in contrast to the other promoters (Al_2O_3, MgO, CaO) TiO_2 had a maximum in the volume of chemisorbed CO at about 0.5 g atom cation per 100 g atom Fe.

BRIEF SUMMARY OF THE INVENTION

The main object of the present invention was to develop a catalyst for ammonia synthesis with improved activity.

Higher activity could be utilized in two ways: High reaction rate at high ammonia concentrations or higher activity at low temperatures.
The catalyst activity may be expressed as a rate constant in a rate equation for the synthesis reaction. The rate is a function of temperature, pressure and the composition of the gas, and decreases rapidly with increasing ammonia concentrations. Thus a large fraction of the catalyst volume in a synthesis converter will have ammonia concentration approaching the exit concentration. Hence, to improve the efficiency, increasing the reaction rate at high ammonia concentration is of perticular interest.
As the equilibrium temperature decreases with increasing ammonia concentration, searching higher conversion by increasing the reaction rate at high ammonia concentration also means searching a catalyst with higher activity at low temperature.

The inventors have manufactured a lot of samples of iron oxide based catalysts with various promoters in various concentrations to improve the catalyst activity. In addition to the conventional promoters mentioned above, new promoters have been tested, in particular cobalt- and titanium oxide.

The inventors found that the reaction rate at high ammonia concentration increased with 10-20% when both cobalt- and titanium oxide were used as promoters together with the conventional promoters.

The most preferred catalyst was achieved when the concentration of cobalt was between 0.1% and 3.0 % by weight of metal and when the concentration of titanium was between 0.1 % and 1.0 % by weight of metal.

The atomic ratio Fe^{2+}/Fe^{3+} was between 0.5-0.65.

The present invention will thus in its widest scope comprise a catalyst for the synthesis of ammonia from hydrogen and nitrogen consisting of iron oxides and promoters where the promoters comprise oxides of both cobalt and titanium in addition to Al, K, Ca and Mg oxides.

The invention will be further explained in the example.

DETAILED DESCRIPTION OF THE INVENTION

Example:

Samples were made by mixing iron ore, mainly magnetite, Fe_3O_4, with promoters. These mixtures were then melted in a ceramic crucible in a laboratory furnace. The temperature was kept at about 1600 °C. The ratio of Fe^{2+}/Fe^{3+} was adjusted to within 0.5 to 0.65 by adding Fe-metal to the melt.
The melt was then poured into an iron crucible and cooled.

The concentration of the promoters in the samples varied as shown in table I:

Table I:

K	0.4 - 0.5
Ca	1.6 - 1.8
Mg	0.3 - 0.5
Al	1.5 - 1.8
Co	0.10- 3.00
Ti	0.14- 0.95

The balance being iron oxides with natural impurities. The cooled samples were crushed and sieved to 0.4 - 0.63 mm particles for testing in a microreactor.